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**(54) Sound insulating thermoplastic resin compositions**

(57) A thermoplastic resin composition for use in sound insulators comprising a polyolefin, an ethylene- $\alpha$ -olefin or monovinyl aromatic hydrocarbon-conjugated diolefin copolymer rubber, process oil and an inorganic filler, particularly when the filler is present in large quantities, has high flexibility, heat resistance and workability characteristics, such as melt-flow. These compositions may be molded in

standard equipment such as injection or extrusion molding.

Preferably the composition includes as the polyolefin component polypropylene or ethylene-propylene block copolymers; as the ethylene- $\alpha$ -olefin rubber component ethylene-propylene copolymers including ethylene-propylene diene terpolymers, for example, ethylene-propylene ethylidenenorbornene rubber; as the monovinyl aromatic hydrocarbon-conjugated diolefin copolymer rubber component, random or block copolymers of styrene butadiene rubber.

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## SPECIFICATION

## Thermoplastic resin compositions

This invention relates to a thermoplastic resin composition, to sound insulators comprising that composition and to a method for their preparation.

5 It is well known to add an inorganic filler into a polyolefin in order to improve its physical properties and lower its cost. With an increase in its content of an inorganic filler, however, a polyolefin has hitherto shown a tendency to increase its density and modulus of elasticity, and lose its flexibility, and has not been suitable for molding a product having both a high density and a high degree of flexibility. In order to obtain a resin having a high content of an inorganic filler, and yet a high degree of flexibility, it is generally known to use, for example, an ethylene-vinyl acetate copolymer, an ethylene-ethyl acrylate copolymer, or an ethylene- $\alpha$ -olefin copolymer in the form of rubber as base material. In order to obtain a highly flexible product from an ethylene-vinyl acetate, or ethylene-ethyl acrylate copolymer, however, it is necessary to use a copolymer having a high proportion of vinyl acetate or ethyl acrylate, which results in a composition having a low melting point, and hence, low heat resistance. A composition containing a high proportion of an ethylene- $\alpha$ -olefin copolymer in the form of rubber is also low in heat resistance, since the rubber per se has a low melting or softening point. 10 15

It has now been found that a particular improved thermoplastic resin composition may contain a high proportion of an inorganic filler, and yet retain high flexibility.

According to this invention, there is thus provided a thermoplastic resin composition comprising (A) a polyolefin, (B) an ethylene- $\alpha$ -olefin, or monovinyl aromatic hydrocarbon-conjugated diolefin copolymer in the form of rubber, (C) process oil and (D) an inorganic filler. 20

The composition obtained by adding an ethylene- $\alpha$ -olefin, or monovinyl aromatic hydrocarbon-conjugated diolefin copolymer rubber, and process oil into a polyolefin retains flexibility even if a large quantity of an inorganic filler is added thereto, and provides a thermoplastic resin composition having high degrees of heat resistance and moldability. 25

In preferred embodiments, the composition comprises 5 to 400 parts by weight of (B) for 100 parts by weight of (A), and 5 to 100 parts by weight of (C) and 10 to 3,200 parts by weight of (D) for a total of 100 parts by weight of (A) and (B).

For the purpose of this invention, the polyolefin may be an  $\alpha$ -olefin homopolymer having 2 to 6 carbon atoms, or a copolymer comprising primarily said homopolymer. The preferred homopolymer is polypropylene. Examples of these polyolefins include high-density polyethylene, medium- or low-density polyethylene, a crystalline ethylene-propylene copolymer, such as ethylene-propylene block copolymer, a crystalline ethylene-butene-1 copolymer, polypropylene, a crystalline propylene-butene copolymer, poly-butene-1, poly-4-methylpentene-1, an ethylene-vinyl acetate copolymer, and ethylene-ethyl acrylate copolymer, and the mixtures thereof. An ethylene-propylene block copolymer is preferred. 30 35

The ethylene- $\alpha$ -olefin copolymer in the form of rubber may be a bipolymer formed by ethylene and an  $\alpha$ -olefin having at least three carbon atoms, such as propylene, butene-1, pentene-1, hexene-1 and 4-methylpentene-1. It is preferable to use a copolymer between ethylene and propylene or butene-1, and ethylene-propylene rubbers are particularly preferred. The ethylene- $\alpha$ -olefin copolymer rubber is, however, not limited to any such bipolymer, but also includes a terpolymer or other copolymer formed by ethylene, an  $\alpha$ -olefin having at least three carbon atoms, and one or more dienes such as 1,4-hexadiene, 1,5-hexadiene, di-cyclopentadiene, methylenenorbornene, ethyldenenorbornene, cyclooctadiene and methyltetrahydroindene. A preferred terpolymer is an ethylene-propylene-(non-conjugated diene) rubber, particularly where the diene is ethyldenenorbornene, 1,4-hexadiene or dicyclopentadiene. The ethylene- $\alpha$ -olefin copolymer in the form of rubber may have an ethylene content of 20 to 80% by weight, preferably 40 to 70% by weight, and a Mooney viscosity ( $ML_{1+4}$  at 100°C) of 10 to 150, preferably 20 to 120. If a terpolymer or other copolymer is used, it is preferable to use one having a diene content of 5 to 40% by weight. 40 45

The monovinyl aromatic hydrocarbon-conjugated diolefin copolymer in the form of rubber for use according to this invention may be a random or block copolymer formed by a monovinyl aromatic hydrocarbon such as styrene, vinyltoluene, vinylxylene, ethylvinylbenzene, isopropylvinylbenzene, ethylvinyltoluene and vinylanthracene, and a conjugated diolefin having 4 to 6 carbon atoms, such as 1,3-butadiene, isoprene, 1,3-pentadiene, and 2,3-dimethyl-1,3-butadiene, and preferably a random or block copolymer of styrene and 1,3-butadiene. The copolymer may have a monovinyl aromatic hydrocarbon content of 20 to 80% by weight, preferably 30 to 70% by weight, and a Mooney viscosity ( $ML_{1+4}$  at 100°C) of 10 to 150, preferably 20 to 120. 50 55

The composition of this invention may contain 5 to 400 parts, preferably 20 to 200 parts, by weight of the ethylene- $\alpha$ -olefin, or monovinyl aromatic hydrocarbon-conjugated diolefin copolymer in the form of rubber for 100 parts by weight of the polyolefin. Any amount below 5 parts by weight results in a sharp reduction in the amount of the inorganic filler which can be added, while any excess over 400 parts by weight leads to a reduction in the melting point of the composition, and its heat resistance. 60

For the purpose of this invention, any process oil may be used, if it can improve the workability of the rubber by lubricating the rubber molecules for their free movement, lowering internal friction to

reduce generation of heat upon mixture, assisting the dispersion of the filler, and improving the compatibility between the polyolefin and the rubber. It is particularly preferable to use a hydrocarbon having a boiling point of at least 350°C. Examples of such process oil include paraffinic, naphthenic and aromatic petroleum fractions having a high boiling point, but it is particularly preferable to use a process oil composed of a paraffinic or naphthenic high boiling petroleum fraction which is highly compatible with the copolymerized rubber. The composition of this invention may contain 5 to 100 parts, preferably 10 to 50 parts, by weight of the process oil for a total of 100 parts by weight of the polyolefin and the rubber. Any excess over the upper limit is hereinabove mentioned may result in a molded product which is low in strength and tacky, while the use of any amount below the lower limit fails to achieve satisfactory extension of the rubber, and effectively improve the flexibility, melt-flow characteristics and moldability of the product as aimed at according to this invention.

The inorganic filler for use according to this invention may be selected from among metals, metal oxides, metal salts, silicates and silicate minerals, and the like. Examples of the metals include iron, zinc, lead, copper, nickel, chromium, molybdenum and manganese; examples of the metal oxides include zinc oxide, titanium oxide, iron oxide, iron sesquioxide, triiron tetroxide, tin oxide, nickel oxide, chromium oxide, molybdenum oxide and alumina; examples of the metal salts include barium sulfate, lead sulfate, calcium sulfate, lead carbonate, barium carbonate, zinc carbonate, calcium carbonate and magnesium carbonate; and examples of the silicates and silicate minerals include calcium silicate, silica powder, silica sand, silica, mica, asbestos, talc and clay. One or more of these inorganic fillers may be appropriately selected to suit the use of the molded product to be obtained. While there is no limitation in particular to the particle size which is not greater than 150  $\mu$ , preferably not greater than 100  $\mu$ , in order to ensure the ease of the mixing and molding operations and protect the molding apparatus against any damage by the filler particles. The composition of this invention may contain 10 to 3,200 parts, preferably 20 to 2,500 parts, by weight of the filler for a total of 100 parts by weight of the polyolefin and the rubber. An amount below 10 parts by weight may fail to produce any effect of the inorganic filler in the molded product, while an excess over 3,200 parts by weight may result in the failure to obtain any satisfactory molded product because of its inferior flexibility and melt-flow characteristics. If the amount of the filler is expressed in parts by volume, it is preferable to employ not more than 200 parts by volume of the inorganic filler for a total of 100 parts by volume of the polyolefin, the copolymerized rubber and the process oil.

The thermoplastic resin composition of this invention may be used as a sound insulator, and for this application it is desirable to use a filler having as high a density as possible, and incorporate filler in an amount permitting the composition to have a density of at least 1.5 gm/cm<sup>3</sup> in order to obtain a satisfactorily high degree of sound insulation, since its sound insulating properties are generally proportional to the logarithm of the product of its surface density (kg/cm<sup>2</sup>) by the frequency. The amount of the filler to be added depends on its density, and if the filler has a density of, for example, 2 gm/cm<sup>3</sup>, it is desirable to employ at least 300 parts by weight of the filler for a total of 100 parts by weight of the polyolefin and the rubber, and it is preferred that the final composition, which also contains the process oil, contain at least 70% by weight of the filler.

The composition of this invention may be prepared by a standard melting and kneading or mixing process. For example, the four constituents of the composition may be melted and kneaded together in an internal mixer such as a Banbury mixer, a roll mixer, or the like but at a temperature which is not lower than the melting point of the polyolefin, but not higher than 300°C. It is also appropriate to mix the constituents preliminarily in a Henschel mixer and knead the mixture through an extruder. The process oil and the copolymerized rubber can be mixed with each other before they are mixed with the other constituents.

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If required, it is possible to incorporate a colouring agent, an antistatic agent, an antioxidant, a lubricant, an ultraviolet light absorber, a heat stabilizer, a surface active agent, or the like into the composition of this invention.

According to this invention, it is thus possible to obtain a thermoplastic resin composition which contains a large quantity of an inorganic filler, and which is superior in flexibility, heat resistance and melt-flow characteristics; this composition can be molded in a customary manner by injection or extrusion molding, or otherwise. The composition of this invention containing a large quantity of an inorganic filler shows a high transmission loss for sound waves as it has a higher density than a polyolefin alone. As it has a lower modulus of elasticity and a higher degree of flexibility than a polyolefin containing the same quantity of an inorganic filler, it does not give rise to any coincidence effect, i.e., the resonance caused by penetration of sound energy into the material through one surface thereof, and emission thereof through the opposite surface. The composition is moreover easy to work.

These and other advantages of the composition according to this invention render it very suitable as sound insulating material. It can, for example, be used as a sheet for protection against noise, roof, wall or floor coverings, aggregates and other building materials, or sound insulating materials for noisy machines, pipelines or the like. In the form of a sheet, it may be laminated on fibre products such as woven or nonwoven fabrics and carpets, sound absorbing materials such as rock wool, glass fibres and foamed polyurethane, or other sound insulating materials such as metal plates, plywood, plaster boards, cement concrete plates and cement mortar plates, or may be sandwiched therebetween to form a triple-layered structure. The composition of this invention may also be laminated on the surface of a structural member in a vehicle, ship, automobile, instrument, machine or the like to damp its vibration and reduce any noise arising therefrom.

The composition of this invention will now be described more specifically with reference to examples, but this invention is not limited by those examples. All parts are shown by weight throughout the examples. The melt index (MI) was obtained in accordance with the testing method of ASTM D—1238 at a temperature of 230°C and a load of 2,160 g for an ethylene-propylene block copolymer and polypropylene, and at a temperature of 190°C and a load of 2,160 g for a high- or low-density polyethylene and an ethylene-vinyl acetate copolymer. The modulus in flexure and the density were obtained in accordance with the testing methods of ASTM D—790 and JIS K—6758, respectively. The melting point was obtained by a differential calorimeter. The flexibility was determined by a feel test on a 3 mm thick sheet of the composition prepared by press forming. In the tables, a double circle means 'very soft'; a single circle, 'soft'; a triangle, 'hard'; and an x, 'very hard'.

#### EXAMPLE 1

An ethylene-propylene block copolymer having an ethylene content of 7% by weight and an MI of 9, ethylene-propylene rubber (EPR) having an ethylene content of 70% by weight and a Mooney viscosity of 70, calcium carbonate having an average particle diameter of 2  $\mu$  and a paraffinic process oil (Kyodo Sekiyu's R—1000) were charged into a Banbury mixer in various proportions as shown in TABLE 1, and kneaded for 10 minutes at a temperature of 190°C to 200°C. After the kneaded mixture was cooled, it was crushed, and the crushed product was pressed at 200°C and 100 kg/cm<sup>2</sup>G to form a sheet of each composition having a thickness of 3 mm. A testpiece for use in the measurement of the flexural modulus and density was prepared from the sheet, and the physical properties of each composition were determined, and its flexibility evaluated. The results are shown in TABLE 1, which also shows results for testpieces prepared from 2 compositions not falling within the invention.

TABLE 1

Run No.	1	2	3	4 Comparative Example	5 Comparative Example
Ethylene-propylene block copolymer (parts)	20	30	10	50	40
EPR (parts)	10	10	5	—	10
Calcium carbonate (parts)	50	50	75	50	50
Process oil (parts)	20	10	10	—	—
Density (g/cm <sup>3</sup> )	1.34	1.35	1.79	1.35	1.35
Flexural Modulus (Kg/cm <sup>2</sup> )	1,350	1,900	3,800	22,600	18,000
Melting point (°C)	153.2	155.1	156.3	160.5	160.1
Flexibility	⊙	○	○	X	Δ

#### EXAMPLE 2

The procedures of Example 1 were repeated for preparing the various compositions shown in TABLE 2, and determining and evaluating their physical properties, except that polypropylene (PP) having an MI of 22 was used instead of the ethylene-propylene block copolymer, that barium sulfate having an average particle diameter of 7  $\mu$  was used instead of calcium carbonate, and that an ethylenevinyl acetate copolymer (EVA) having an MI of 20 and a vinyl acetate content of 28% by weight was used. The results are shown in TABLE 2, again showing results for 2 comparative examples.

TABLE 2.

Run No.	6	7	8	9 Comparative Example	10 Comparative Example
PP (parts)	25	15	10	—	45
EVA (parts)	—	—	—	45	—
EPR (parts)	10	15	20	—	—
Barium sulfate (parts)	55	55	55	55	55
Process oil (parts)	10	15	15	—	—
Density (g/cm <sup>3</sup> )	1.60	1.59	1.58	1.61	1.61
Flexural Modulus (Kg/cm <sup>2</sup> )	2,100	1,025	750	4,000	20,000
Melting point (°C)	155.4	153.8	150.0	65	161.5
Flexibility	O	⊙	⊙	O	X

## EXAMPLE 3

Various compositions were prepared by kneading various proportions, as shown in TABLE 3, of polypropylene (PP) having an MI of 15, an ethylene-propylene block copolymer having an ethylene content of 7% by weight and an MI of 9, high-density polyethylene (HDPE) having a density of 0.096 and an MI of 0.3, low-density polyethylene (LDPE) having a density of 0.912 and an MI of 5, an ethylene-vinyl acetate copolymer (EVA) having an MI of 3 and a vinyl acetate content of 10% by weight, an ethylene-propylene, ethyldenenorbornene terpolymer (EPDM) having a propylene content of 40% by weight, an ethyldenenorbornene content of 15% by weight and a Mooney viscosity of 105, ethylene-propylene rubber (EPR) having an ethylene content of 70% by weight and a Mooney viscosity of 70, talc having an average particle diameter of 12  $\mu$  and a paraffinic process oil (Witco Chemical's product known as Protol). The physical properties of each composition were determined and evaluated in accordance with the same procedures as employed in Example 1. The results are shown in TABLE 3, again including the results for 2 comparative examples.

TABLE 3

Run No.	11	12	13	14
PP (parts)	15	10	10	10
Ethylene-propylene block copolymer (parts)	—	—	—	—
EDPE (parts)	—	5	—	—
LDPE (parts)	—	—	5	—
EVA (parts)	—	—	—	5
EPDM (parts)	10	10	10	10
EPR (parts)	—	—	—	—
Talc (parts)	60	60	60	60
Process oil (parts)	15	15	15	15
Density (g/cm <sup>3</sup> )	1.53	1.53	1.53	1.53
Flexural Modulus (Kg/cm <sup>2</sup> )	4,000	3,500	3,000	2,300
Melting point (°C)	155.1	154.1	153.2	151.4
Flexibility	O	O	O	O

TABLE 3 (Continued)

Run No.	15 Comparative Example	16	17 Comparative Example
PP (parts)	40	10	—
Ethylene-propylene block copolymer (parts)	—	—	25
EDPE (parts)	—	—	—
LDPE (parts)	—	—	—
EVA (parts)	—	—	—
EPDM (parts)	—	—	—
EPR (parts)	—	5	—
Talc (parts)	60	75	75
Process oil (parts)	—	10	—
Density (g/cm <sup>3</sup> )	1.53	1.68	1.69
Flexural Modulus (Kg/cm <sup>2</sup> )	50,000	4,500	60,000
Melting point (°C)	161.5	158.4	162.3
Flexibility	X	O	X

## EXAMPLE 4

Various compositions were prepared and evaluated by repeating the procedures of Example 1, except that styrene-butadiene rubber having a styrene content of 40% by weight and a Mooney viscosity of 24 was used instead of the ethylene-propylene rubber, zinc oxide having an average particle diameter of 1  $\mu$  or below instead of the calcium carbonate, and Witco Chemical's Protol instead of the Kyodo Sekiyu's paraffinic process oil, and that the constituents were kneaded by a roll mixer for 10 minutes at about 200°C in various proportions as shown in TABLE 4. The results are shown in TABLE 4, including results for 3 comparative examples.

TABLE 4

Run No.	18	19	20 Comparative Example	21 Comparative Example	22 Comparative Example
Ethylene-propylene block copolymer (parts)	20	30	5	—	50
SBR (parts)	10	10	45	50	—
Zinc oxide (parts)	100	100	100	100	100
Process oil (parts)	20	10	—	—	—
Density (g/cm <sup>3</sup> )	2.02	2.03	2.03	2.03	2.04
Flexural Modulus (Kg/cm <sup>2</sup> )	1,500	2,000	3,600	2,900	19,800
Melting point (°C)	152.9	154.1	— (Note)*	— (Note)*	160.3
Flexibility	⊙	O	Δ	O	X

(Note)\*: The high content of non-crystalline styrene-butadiene block copolymer rubber prohibits measurement of the melting point, and renders the composition inferior in heat resistance.

## EXAMPLE 5

Various compositions were prepared and evaluated in physical properties by repeating the procedures of Example 1 except that an ethylene-propylene-ethylidene-norbornene terpolymer (EPDM) having a propylene content of 40% by weight, an ethylenenorbornene content of 15% by weight and a Mooney viscosity of 105 was used instead of the ethylene-propylene rubber, and iron powder having an average particle diameter of 90  $\mu$  or iron oxide having an average particle diameter of 1  $\mu$  instead of the calcium carbonate, and that the constituents were kneaded in the various proportions shown in TABLE 5. The results are shown in TABLE 5; including results for 2 comparative examples.

TABLE 5

Run No.	23	24 Comparative Example	25	26 Comparative Example
Ethylene-propylene block copolymer (parts)	20	40	20	40
EPDM (parts)	10	—	10	—
Iron powder (parts)	120	120	—	—
Iron oxide (parts)	—	—	120	120
Process oil (parts)	10	—	10	—
Density (g/cm <sup>3</sup> )	2.66	2.68	2.33	2.35
Flexural Modulus (Kg/cm <sup>2</sup> )	2,500	24,000	2,200	21,000
Melting point (°C)	155.5	161.4	154.8	161.5
Flexibility	O	X	O	X



As is obvious from the foregoing description, the composition of this invention has a lower modulus of elasticity and a higher degree of flexibility than a composition comprising a polyolefin and an inorganic filler despite their substantially equal density, and is superior in heat resistance to a composition comprising a low-crystalline or non-crystalline resin and an inorganic filler. Moreover, the composition of this invention is superior in workability, as it has been extrusion molded into a sheet having an excellent surface which is free from any roughness or unevenness. 5

#### CLAIMS

1. A thermoplastic resin composition comprising (A) a polyolefin, (B) an ethylene- $\alpha$ -olefin or monovinyl aromatic hydrocarbon-conjugated diolefin copolymer in the form of a rubber, (C) process oil and (D) an inorganic filler. 10
2. A composition of matter according to Claim 1, wherein said polyolefin is an  $\alpha$ -olefin homopolymer having 2 to 6 carbon atoms, or a copolymer comprising primarily the homopolymer with mixtures of similar homopolymers.
3. A composition according to Claim 2, wherein said homopolymer is polypropylene.
4. A composition according to Claim 2, wherein said copolymer is an ethylenepropylene block copolymer. 15
5. A composition according to any of Claims 1—4, wherein said ethylene- $\alpha$ -olefin copolymer rubber is an ethylene-propylene rubber or ethylene-propylene-(non-conjugated diene) rubber.
6. A composition according to Claim 5, wherein said non-conjugated diene is ethylidenenorbornene, 1,4-hexadiene or dicyclopentadiene. 20
7. A composition according to Claim 6, wherein said diene is present from about 5 to about 40% by weight of said ethylene-propylene-(non-conjugated diene) rubber.
8. A composition according to any of Claims 1—4, wherein said monovinyl aromatic hydrocarbon-conjugated diolefin copolymer rubber is a random or block copolymer rubber.
9. A composition according to Claim 8, wherein said copolymer is styrenebutadiene rubber. 25
10. A composition according to any of Claims 1—9, including 5 to 400 parts by weight of (B) for 100 parts by weight of (A).
11. A composition according to any of Claims 1—10, including 5 to 100 parts by weight of (C) for a total 100 parts by weight of (A) and (B).
12. A composition according to any of Claims 1—11, including 10 to 3,200 parts by weight of (D) for a total of 100 parts by weight of (A) and (B). 30
13. A sound insulator comprising a thermoplastic resin composition comprising (A) a polyolefin, (B) an ethylene- $\alpha$ -olefin, or monovinyl aromatic hydrocarbon-conjugated diolefin copolymer rubber, (C) process oil and (D) an inorganic filler.
14. A sound insulator according to Claim 13 wherein the density of said composition is at least 1.5 gm/cm<sup>3</sup>. 35
15. A sound insulator according to Claim 14 wherein the composition is as claimed in any of Claims 2—12.
16. A method for producing a sound insulator comprising a thermoplastic resin composition comprising (A) a polyolefin, (B) an ethylene- $\alpha$ -olefin, or monovinyl aromatic hydrocarbon-conjugated diolefin copolymer rubber, (C) a process oil and (D) an inorganic filler wherein components (A), (B), (C) and (D) are mixed together at a temperature which is not lower than the melting point of said polyolefin, but not higher than 300°C. 40
17. A method according to Claim 16 wherein the composition is as claimed in any of Claims 2—12 and 14. 45

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